2013

Scale-up of Bubbling Fluidized Bed Reactors with Vertical Internals: A New Approach Accounting for Chemistry and Hydrodynamics

Simon Maurer
Paul Scherrer Institut, Switzerland

Martin Rüdisüli
Paul Scherrer Institut, Switzerland

Tilman J. Schildhauer
Paul Scherrer Institut, Switzerland

Serge M. A. Biollaz
Paul Scherrer Institut, Switzerland

J. Ruud van Ommen
Delft University of Technology, Netherlands

Scale-up of bubbling fluidized bed reactors with vertical internals:

A new approach accounting for chemistry and hydrodynamics

Simon Maurer[a], Martin Rüdisüli[a], Tilman J. Schildhauer[a]*, Serge M. A. Biollaz[a],

J. Ruud van Ommen[b]

[a]Paul Scherrer Institut, 5232 Villigen PSI, Switzerland

[b]Delft University of Technology, Julianalaan 136, 2628 BL Delft, The Netherlands

*T: +41 56 310 27 06; E: tilman.schildhauer@psi.ch

ABSTRACT

The full set of scaling law derived by Glicksman [1] allows the hydrodynamic scale-up of fluidized bed reactors. In case of catalytic reactors, changing the catalyst particle diameter during scale-up may have consequences for the catalyst activity, selectivity and deactivation behavior. For fluidized bed reactors with vertical internals, a new scale-up approach is proposed and tested that helps to avoid this dilemma.

INTRODUCTION

Since the first fluidized bed reactor was erected in 1921 by Fritz Winkler, a large knowledge about the scale dependencies of mass transfer, residence time etc. in fluidized beds was formed Squires [2]. Usually, a selection of dimensionless numbers, e.g. the full set of scaling relations derived by Glicksman, is applied for scale-up Rüdisüli et al. [3]. Today scale-up failures like the well-known that happened in Brownsville (TX, USA) in 1950 can normally be avoided. In Brownsville, a Fischer-Tropsch fluidized bed reactor was directly scaled from an inner diameter of 0.305 m to a 5 m reactor and as consequence of the improper and large scale-up, the residence time and conversion of the reactor was completely changed. The reason was that the bubble diameter in the lab scale reactor exceeded a critical value of 2/3 times the reactor diameter and slugging occurred Hovmand & Davidson [4], whereas in the industrial scale reactor, the bubbling regime was maintained and the bubbles rose much faster leading to a smaller residence time and conversion.

Glicksman's full set of scaling relations is often considered to predict the scale-up of fluidized bed processes. More recent studies however claimed that Glicksman's scaling laws led to mismatches in different scales, especially for larger gas velocities Sanderson & Rhodes, van Ommen et al. [5, 6]. Foscolo et al. also criticized the lack of a particle pressure term, which shall help to account for homogeneous fluidization [7]. Additionally, in case of catalytic reactors, changing the catalyst particle diameter during scale-up may have consequences for the catalyst activity, selectivity and deactivation behavior. The mass transfer limitations could be changed, leading to a significantly different chemical behavior of the scaled reactor, with respect to selectivity and catalyst deactivation.

This paper demonstrates the advantages of the sectoral scale-up approach to scale fluidized bed reactors with vertical internals Rüdisüli et al. [8] and tries to define its limitations. To this end, the restrictions of Glicksman's scale-up law are theoretically derived and a set of experiments, using pressure fluctuation measurements is

performed, to study the hydrodynamic behavior with different numbers of vertical tubes. The sectoral scale-up approach can be applied for all reactors with vertical internals, e.g. heat exchanger tubes. It is based on the full set of Glicksman's scale-up laws; however, the reactor diameter is replaced by the hydraulic diameter of the vertical internals. The new approach leads to a constant catalyst particle size at all scales and is considered as a new possibility to scale both, hydrodynamics and chemistry in a proper way.

THEORY

In 1984, Glicksman derived the most widely used scale-up approach for fluidized beds based on the conservation of mass and momentum in a nondimensional form Glicksman [1], for both particles and fluid. The derivation of the governing equations was performed assuming an incompressible fluid and omitting all inter-particle forces apart from collision forces. The dimensionless parameters derived by Glicksman are, from left to the right, the Reynolds number, the Froude number, the gas-solid density ratio, the bed geometry ratio, the sphericity of the particles and the particle size distribution, Eq. 1.

$$\frac{u_0 \rho_g D}{\eta}, \frac{u_0^2}{g\ D}, \frac{\rho_g}{\rho_p}, \frac{D}{H}, \frac{d_p}{D}, \phi, psd \qquad \textbf{Eq. 1}$$

Glicksman concluded in his work that the motion of fluid and particles can be properly scaled for fluidized beds. However, for catalytic gas conversion processes in fluidized beds, the conversion and yield is commonly a function of the mass transfer (limitations), selectivity and activity of the catalyst particles involved. These parameters are not directly taken into account for Glicksman's scale-up law and may be changing during scale-up. Even if for scaled catalyst particle size, the selectivity and activity are the same, the mass transfer is not. In bubbling fluidized beds, three different mass transfer resistances can be distinguished. That are, the mass transfer resistance between bubble and dense phase, the external mass transfer around a catalyst particle and the internal mass transfer resistance inside the catalyst particle.

In bubbling fluidized beds, the mass transfer between bubble and dense phase usually limits the overall reaction rate. This is why scale-up of bubbling fluidized beds is more challenging compared to other fluidization regimes, where the external, internal mass transfer and/or chemistry control the reaction rate. In Eq. 2, Sit & Grace [9], the overall mass transfer coefficient from the bubble to the dense phase is depicted as sum of a convective and diffusive mass transfer term. It is assumed, that the mean bubble rise velocity is larger than the gas velocity in the dense phase. Similar mass transfer correlations neglecting the convective transport were published by Kunii & Levenspiel, Davidson & Harrison [10, 11].

$$k_c = \frac{u_{mf}}{3} + \left(\frac{4\ D_g \varepsilon_{mf}\ u_b}{\pi\ d_b} \right)^{1/2} \qquad \textbf{Eq. 2}$$

As can be seen in Eq. 2, the convective mass transfer term is a function of the minimal fluidization velocity, which is normally derived from the well known Ergun equation Ergun [12]. Both, the minimal fluidization velocity and therefore the mass transfer coefficient are depending on the particle size and are changed during scale-up. The diffusive term is a function of the mean bubble diameter and velocity, which depend on the bed material used. When different bubble size correlations are analyzed, approximately 50% show a direct dependence on the particle diameter e.g. Kato & Wen, Yasui & Johanson, Park et al. [13, 14, 15] and nearly all show an indirect

dependency on the minimal fluidization velocity. However, when Glicksman's full set of scaling law is applied, almost all mass transfer correlations are scaled in another way.

A similar problem with scale-up can be found, when the external mass transfer coefficient around a catalyst particle is analyzed. The external mass transfer coefficient in fluidized beds can be calculated according to Eq. 3, derived from numerous published mass transfer measurements Dwivedi & Upadhyay [16]. The mass transfer is a function of the particle Reynolds number, the superficial gas velocity, the void fraction and the Schmidt number. Again, the mass transfer is usually changed during scale-up and the changes may differ for different mass transfer correlations.

$$k_c = \frac{0.4548\ \mathrm{Re}_p^{-0.4069}\ u_0}{\varepsilon\ Sc^{2/3}} \qquad \textbf{Eq. 3}$$

The internal mass transfer limitation of a catalyst particle is also a function of its diameter and may change during scale-up.

An easy and direct way to avoid these dependencies and to take all mass transfer limitations as well as the change in selectivity and activity of a catalytic particle into account is to keep the same catalyst particle size for the different scales. This can be done with the sectoral scale-up approach for all fluidized beds with vertical internals. The sectoral scale-up approach basically replaces the absolute bed diameter in Glicksman's dimensionless parameters with the hydraulic diameter of the bed Rüdisüli et al. [8]. In Figure 1, the new sectoral and traditional scale-up approach are visualized.

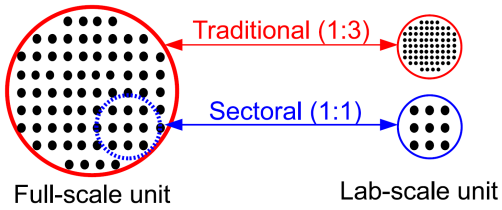

Figure 1 Schematic principle of the traditional and new sectoral scale-up approach, adapted from Rüdisüli et al. [8]

EXPERIMENTAL

The hydraulic experiments were conducted in a lab-scale glass column called Glas15, with 14.5 cm inner diameter and vertical internals. The vertical internals had a diameter of 1 cm, 0.9 cm tube spacing and a squared arrangement. In Figure 2 left-hand side, the complete setup can be seen. The red ports in front of the column are the flanges to connect the Kistler sensors (piezo-electric pressure transducer, type 7261) for pressure fluctuation measurements.

Different u-shaped profiles were introduced to the setup (Figure 2, right-hand side) to study the bubble size with 19, 28 and 35 vertical tubes at different gas velocities. The u-shaped metal plates were inserted in the Glas15 fluidized bed reactor and sealed to the outer reactor wall by closed pore foam. The gas tight sealing of the u-shape profiles was tested by comparing pressure drop versus gas velocity for each alignment with the values measured in the open, circular bed. That ensures a defined gas-flow inside the rectangular area, where the measurement shall take place. With the measurements, the minimal number of vertical tubes and maximal bubble to reactor diameter for successful scale-up shall be determined.

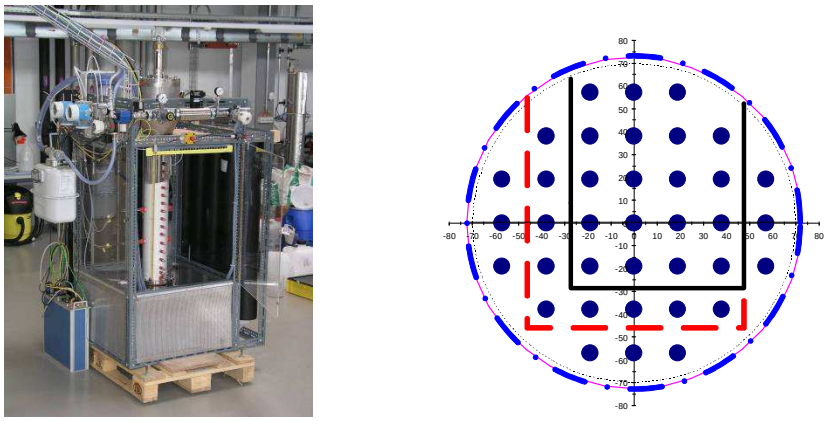

Figure 2 Left-hand side, lab scale glass column with red pressure fluctuation ports in the front, right hand side top view of the column with different u-profiles to reduce the number of vertical tubes

For all experiments, porous γ-aluminum oxide powder, called NWA, was used. The particle characterization has been reported by Rüdisüli et al. [17]. The sauter mean diameter of the powder was determined by a sieve analysis and laser diffraction measurements to be 289 µm, the sphericity was obtained by a SEM image to be 0.59. The dense phase voidage was calculated with a correlation published by Wen & Yu [18] to be 0.5. The particle density is provided by the manufacturer to be 1350 kg/m^3 and the internal angle of friction was calculated by applying the "bin-flow" method of Zenz & Othmer [19] to be 33.3°. The particles are in the intermediate range between Geldart A and B Geldart [20, 21].

Six Kistler sensors were simultaneously mounted at the Glas15 for pressure fluctuation measurements. One served as reference sensor in the windbox below the distributor plate, four sensors were taken for measurements inside the bed and one sensor directly above the bed surface. The sensors were mounted at 27, 81, 243, 350, 450 mm. With pressure fluctuation measurements, a value proportional to the average bubble diameter can be obtained from the incoherent output signal of a sensor in the bed Chilekar et al. [22]. Additionally, the power spectral density can be applied to investigate proper scale-up Fitzgerald & Crane, Nicastro & Glicksman, Stein et al., Di Felice et al. [23, 24, 25, 26].

RESULTS

From the incoherent output of the pressure fluctuation measurement, the characteristic length σ, which is proportional to the average bubble diameter, is obtained. It is plotted in function of the measurement position in the bed, see Figure 3. The measurements have been conducted for three different ratios between gas velocity and minimal fluidization velocity u/u_{mf}.

In Figure 3, the bubble characteristics are approximately equal for 28 and 37 tubes, however for 19 tubes, the situation differs. The hydrodynamic behavior of the column seems to be changed for the lowest number of vertical tubes, which means that the characteristic length of the bubble is changed.

At least two reasonable explanations are possible. The first explanation is that the outer wall is getting more important for the column with 19 tubes and the smaller cross-sectional area. This would lead to an increased bubble size in the upper part of the fluidized bed, because the probability for bubble coalescence is increased. The increase in bubble size can be seen in the column with 19 vertical tubes only, but for all gas velocities.

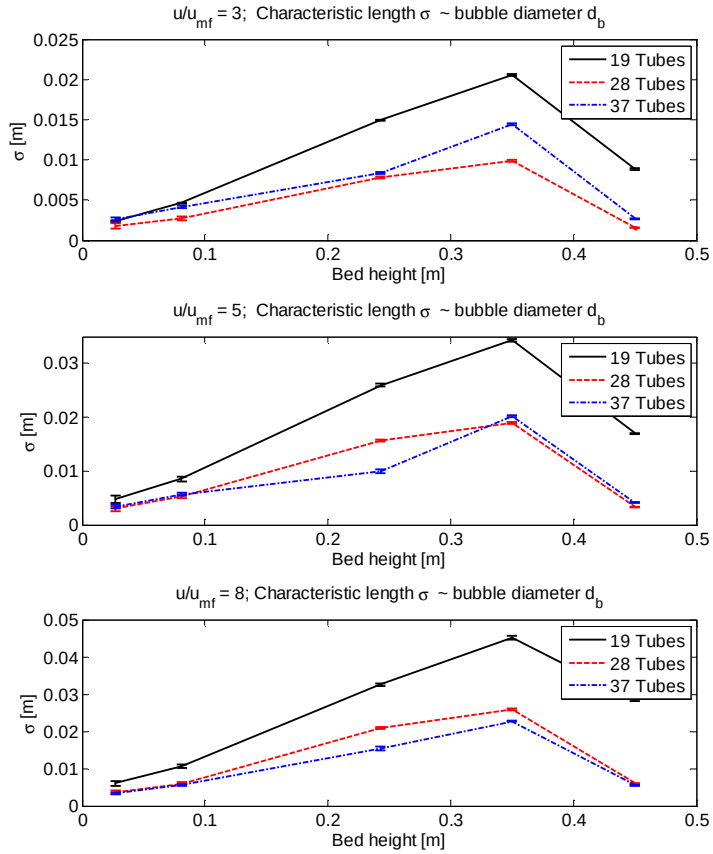

Figure 3 Characteristic length-scale for different gas velocities

However, this increase in bubble size (or characteristic length) may also be explained by the decreased cross-sectional area and the measurement principle itself. The decrease in total cross-sectional area could in general lead to a higher pressure signal for the same bubble size, because less area or volume in a single element is available, leading to a larger pressure signal for the same bubble and therefore to an increase in the calculated characteristic length.

A similar picture can be seen, if the incoherent output (IOP) of the pressure fluctuation measurements is compared, Figure 4. At all bed heights and gas velocities, the IOP for 28 and 37 tubes is approximately equal, although, the cross sectional area is decreased. The IOP for 19 tube arrangement starts to differ. For 27 and 81 mm measurement height, the values between different numbers of tubes differ only slightly and the shape of the IOPs stayed approximately the same. However, for larger measurement heights the differences are increased and also the slope and shape of the IOPs are completely changed. It seems like the wall effects are getting dominant, especially in the upper regions of the fluidized bed.

From the optical observation of the glass column, slugging was observed in the upper region of the fluidized bed for 19 tubes at all gas velocities. For gas velocities of u/u_{mf} = 5 and 8 and the 27 tube arrangement it seemed like the bed was in a transition regime between slugging and bubbling, because a minimal change can be seen in the IOPs and by optical observation larger bubbles appeared from time to time.

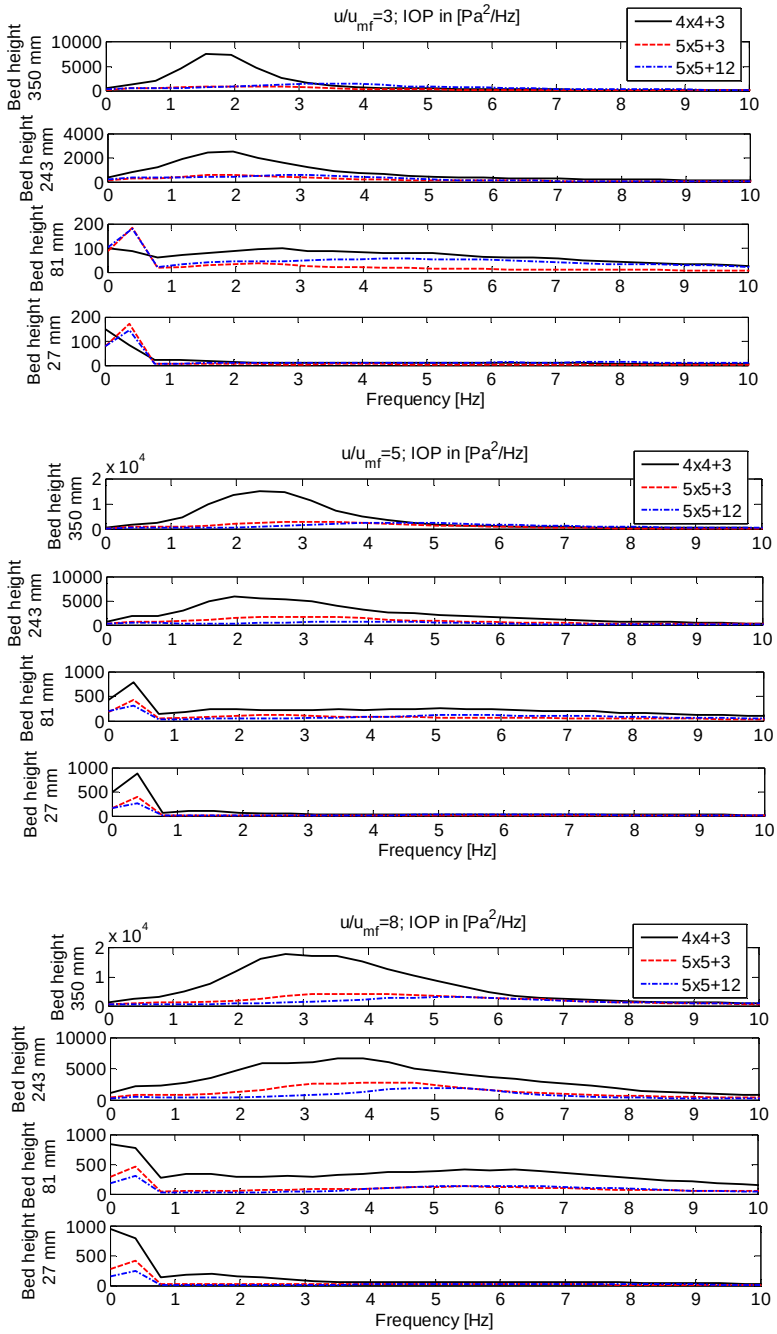

Figure 4 Incoherent output of the pressure fluctuation measurement at different gas velocities and measurement heights

CONCLUSIONS

The approach described in this paper shows obvious advantages for the scaling of fluidized beds with catalytic gas conversion processes. The complexity of the scale-up is significantly decreased. Especially for catalytic bubbling fluidized beds with vertical internals, efficient scale-up seems to be now reasonable. All usually scale-dependent parameters, such as mass transfer, selectivity and activity, are kept constant. The only disadvantage seen right now is the constant bed height at all scales. To proof the concept, a first series of comparison measurements was conducted in a glass column

with different cross sectional areas but constant hydraulic diameter of the internals. Pressure fluctuation measurements were applied to study the hydrodynamics for three different gas velocities and cross sectional areas. From a naive point of view one could argue that at least 28 tubes are needed in a column of 90 mm in diameter. However, as discussed, the measured characteristic length can be reasonable explained in two different ways. Therefore, a second series of measurements with optical probes will be conducted with the same reactor and configuration to compare mean bubble sizes, velocities and their distributions, and to increase the validity of the conclusions made. Nevertheless, the new method shall enable industry to design and optimize bubbling fluidized bed reactors with vertical internals with reduced costs and development time.

NOTATION

D	fluidized bed diameter	ε	void fraction
d_b	average bubble diameter	ε'	fixed bed porosity
D_g	gas diffusion coefficient	ε_{mf}	porosity at u_{mf}
d_p	particle diameter	η	dynamic viscosity
g	gravity	ρ_g	gas density
H	fluidized bed height	ρ_p	particle density
k_c	mass transfer coefficient	Φ	sphericity
psd	particle size distribution		
Re_p	particle Reynolds number		
Sc	Schmidt number		
u_0	superficial gas velocity		
u_b	swarm bubble rise velocity		
u_{mf}	minimal fluidization velocity		

REFERENCES

[1] Glicksman, L. R.: Scaling relationships for fluidized beds. Chemical Engineering Science. 39(9): 1373 – 1379, 1984.

[2] Squires, A. M.: Contributions toward a history of fluidization. In Proceedings of the Joint Meeting of Chemical Engineering Society of China and AIChE. Beijing, China, Chemical Industry Press, 1982, pp 322–353.

[3] Rüdisüli, M., Schildhauer, T. J., Biollaz, S. M., and van Ommen, J. R.: Scale-up of bubbling fluidized bed reactors - a review. Powder Technology. 217(0): 21–38, 2012.

[4] Hovmand, S. and Davidson, J. F. Fluidization: Pilot plant and laboratory scale fluidized reactors at high gas velocities; the relevance of slug flow, chapter 5. Academic Press, New York, 1971.

[5] Sanderson, J. and Rhodes, M.: Bubbling fluidized bed scaling laws: Evaluation at large scales. AIChE Journal. 51(10): 2686–2694, 2005.

[6] van Ommen, J. R., Teuling, M., Nijenhuis, J., and van Wachem, B. G. M.: Computational validation of the scaling rules for fluidized beds. Powder Technology. 163(1-2): 32–40, 2006.

[7] Foscolo, P. U., Gibilaro, L. G., and Di Felice, R.: Hydrodynamic scaling relationships for fluidisation. Applied Scientific Research. 48: 315–328, 1991.

[8] Rüdisüli, M., Schildhauer, T. J., Biollaz, S. M., and van Ommen, J. R.: Evaluation of a sectoral scaling approach for bubbling fluidized beds with vertical internals. Chemical Engineering Journal. 197(0): 435 – 439, 2012.

[9] Sit, S. and Grace, J.: Effect of bubble interaction on interphase mass transfer in gas fluidized beds. Chemical Engineering Science. 36(2): 327 – 335, 1981.

[10] Kunii, D. and Levenspiel, O.: Bubbling bed model. model for flow of gas through a fluidized bed. Industrial & Engineering Chemistry Fundamentals. 7(3): 446–452, 1968.

[11] Davidson, J. F. and Harrison, D.: Fluidized particles. New York, Cambridge University Press, Cambridge, 1st edition, 1963.

[12] Ergun, S.: Fluid flow through packed columns. Chemical Engineering Progress. 48(2): 89–94, 1952.

[13] Kato, K. and Wen, C. Y.: Bubble assemblage model for fluidized bed catalytic reactors. Chemical Engineering Science. 24(8): 1351–1369, 1969.

[14] Yasui, G. and Johanson, L. N.: Characteristics of gas pockets in fluidized beds. AIChE Journal. 4(4): 445–452, 1958.

[15] Park, W. H., Kang, W. K., Capes, C. E., and Osberg, G. L.: The properties of bubbles in fluidized beds of conducting particles as measured by an electroresistivity probe. Chemical Engineering Science. 24(5): 851–865, 1969.

[16] Dwivedi, P. N. and Upadhyay, S. N.: Particle-fluid mass transfer in fixed and fluidized beds. Industrial & Engineering Chemistry Process Design and Development. 16(2): 157–165, 1977.

[17] Rüdisüli, M. Characterization of rising gas bubbles in fluidized beds by means of statistical tools. PhD thesis, ETH Zürich, 2012.

[18] Wen, C. Y. and Yu, Y. H.: A generalized method for predicting the minimum fluidization velocity. AIChE Journal. 12(3): 610–612, 1966.

[19] Zenz, F. A. and Othmer, D. F.: Fluidization and fluid-particle systems. Reinhold, 1960.

[20] Geldart, D.: The effect of particle size and size distribution on the behaviour of gas-fluidised beds. Powder Technology. 6: 201–215, 1972.

[21] Geldart, D.: Types of gas fluidization. Powder Technology. 7: 285–292, 1973.

[22] Chilekar, V. P., Warnier, M. J. F., van der Schaaf, J., Kuster, B. F. M., Schouten, J. C., and van Ommen, J. R.: Bubble size estimation in slurry bubble columns from pressure fluctuations. AIChE Journal. 51(7): 1924–1937, 2005.

[23] Fitzgerald, T. J. and Crane, S. D.: Cold fluidized bed modeling. In Proceedings of 5th International Conference on Fluidized Bed Combustion. volume III, Atlanta, GA, USA, 1980, pp 815–820.

[24] Nicastro, M. T. and Glicksman, L. R.: Experimental verification of scaling relationships for fluidized bed. Chemical Engineering Science. 39(9): 1381–1391, 1984.

[25] Stein, M., Ding, Y. L., and Seville, J. P. K.: Experimental verification of the scaling relationships for bubbling gas-fluidised beds using the PEPT technique. Chemical Engineering Science. 57(17): 3649–3658, 2002.

[26] Di Felice, R., Rapagnà, S., and Foscolo, P. U.: Dynamic similarity rules: Validity check for bubbling and slugging fluidized beds. Powder Technology. 71(3): 281–287, 1992.